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# **Special Collection:**

Advances and Best Practices in Boron-based paleo-CO2 reconstruction

### **Key Points:**

- Inverting cloud top XCO<sub>2</sub> using lidar reflection signals above the cloud enhances the utilization of satellite observation data
- Based on passive satellite and model data, the accuracy of atmospheric environment monitoring satellite data inversion is demonstrated
- First use of active remote sensing satellites for assessment of marine carbon uptake

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# MAO ET AL.

# Measurement of CO<sub>2</sub> Column Concentration Above Cloud Tops With a Spaceborne IPDA Lidar

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**Abstract** The Atmospheric Environment Monitoring Satellite (AEMS), launched by China in 2022, was equipped with active remote sensing lidar for carbon monitoring. It adopts the Integrated Path Differential Absorption (IPDA) technology to monitor global  $CO_2$  column concentration (XCO<sub>2</sub>). The calculation of cloud top XCO<sub>2</sub> requires cloud height data. A comparison between SRTM global elevation data and 1,572 nm channel elevation data reveals a coefficient of determination ( $\mathbb{R}^2$ ) of 0.998, with an average deviation of 1.24 m. The cloud top XCO<sub>2</sub> observations are consistent with the OCO-2 and CarbonTracker trends. The ocean carbon uptake rate, assessed by the difference in CO<sub>2</sub> concentration between cloud top and sea surface, is  $-0.319 \text{ mmol/m}^2/\text{h}$ , which is in good agreement with the associated carbon flux data. This demonstrates the great potential of IPDA lidar for remote sensing of cloud top CO<sub>2</sub> and quantifying ocean carbon uptake.

**Plain Language Summary** For global greenhouse gas monitoring, passive remote sensing technology has consistently struggled to balance the reliability and usability of monitoring data in cloudy regions. The AEMS employs 1,572 nm IPDA lidar technology for active remote sensing of global XCO<sub>2</sub>, enabling effective processing and utilization of cloud echo data. In this study, we focused on the concentrations of CO<sub>2</sub> columns using cloud top echoes and performed a preliminary comparison of cloud top XCO<sub>2</sub> results with related data products from the passive satellite OCO-2 and CarbonTracker. By quantifying the difference in CO<sub>2</sub> concentration between two altitude layers above the sea surface, we assessed ocean carbon absorption capacity, and the results demonstrated high reliability. This work highlights the significant advantages of spaceborne IPDA lidar in global CO<sub>2</sub> measurement, cloud echo data processing, and ocean carbon flux assessment, providing valuable data support for climate change research.

# 1. Introduction

 $CO_2$  is the most significant greenhouse gas (GHG) in the atmosphere, contributing approximately 80% to the increase in radiative forcing over the past 5 years (IEA, 2022). The continuous rise in its concentration, primarily driven by human activities, is a key factor in global climate warming. Therefore, high-precision, all-weather, and extensive observations of atmospheric  $CO_2$  concentrations are crucial for advancing carbon reduction efforts, identifying carbon sources and sinks, understanding the carbon cycle, promoting carbon science applications, and supporting global climate change research (Araki et al., 2010; Zhao et al., 2023). Monitoring  $CO_2$  in the lower atmosphere through remote sensing is essential for identifying carbon sources and sinks. Current greenhouse gas monitoring satellites, such as GOSAT, OCO-2, and OCO-3, measure sunlight scattered from the Earth's surface to calculate total column  $CO_2$  content. After decades of development, this technology has become highly accurate. However, passive remote sensing, which relies on sunlight, is limited by solar elevation angles, making it incapable of observations at night and in high-latitude regions. It is also vulnerable to interference from clouds and aerosols, which can obscure slight variations in  $CO_2$  concentration and reduce the reliability of data inversion under cloudy conditions (Chevallier et al., 2014; Liang et al., 2017; Zheng et al., 2023). Approximately two-thirds



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of the Earth's surface is covered by clouds, which further diminishes the effectiveness of passive remote sensing satellite data (Feng et al., 2016). Therefore, compensating for the missing  $CO_2$  information in cloudy regions would significantly enhance the usability of observation data (Baker et al., 2010; Palmer et al., 2019).

In 2007, the U.S. National Research Council proposed the ASCENDS program in its investigative report, which aims to actively detect atmospheric CO<sub>2</sub> emissions during both nighttime and daytime across different seasons (US NRC, 2007). During NASA's ASCENDS airborne experiment activities in 2011, high-resolution measurements of absorption line shapes were conducted using airborne equipment to distinguish echoes from shallow cumulus cloud tops and the ground. This enabled the determination of CO<sub>2</sub> VMR (volume mixing ratio) in the PBL (planetary boundary layer). In subsequent studies, the cloud slicing method was used to derive VMRs for three vertical layers (Ramanathan et al., 2015). The study found that compared to ground-based XCO<sub>2</sub> measurements, the bias in cloud top CO<sub>2</sub> measurements is smaller, but the standard deviation is larger. This is primarily influenced by cloud top roughness and reflectance (Mao et al., 2018). In 2017, the Shanghai Institute of Optics and Fine Mechanics (SIOM) of the Chinese Academy of Sciences conducted a ground-based validation experiment for atmospheric CO<sub>2</sub> measurement using a 1.57 µm IPDA lidar system (Du et al., 2017). Subsequently, SIOM collaborated with NUIST and other institutions to successfully develop a scaled-down airborne version of the spaceborne ACDL system. This system was deployed for airborne CO2 observation experiments (Zhu et al., 2020). In 2018 and 2021, two airborne calibration flights were conducted in Shanhaiguan, a coastal city, and Dunhuang, a desert region, to demonstrate the feasibility and precision limits of the system and its retrieval algorithms under complex geographical conditions (Fan et al., 2024; Wang et al., 2022). Both ground-based and airborne results indicated that the ACDL prototype could achieve sub-ppm accuracy, providing high-precision CO2 measurements on a global scale (Wang, Mustafa, et al., 2021; Zhu et al., 2021). In April 2022, China launched the world's first satellite equipped with CO<sub>2</sub> laser detection capabilities: the Atmospheric Environment Monitoring Satellite (AEMS). Its primary payload, the ACDL system, incorporates an IPDA lidar designed for global CO<sub>2</sub> monitoring. AEMS is designed to measure atmospheric CO<sub>2</sub> with a precision of better than 1 ppm, featuring a land resolution of 50 km and an ocean resolution of 100 km. Using the Total Carbon Column Observing Network (TCCON) as the reference for cross-validation, comparisons between AEMS and TCCON XCO2 observations indicate that ACDL's measurement bias is less than 1 ppm, with a system bias of  $0.1 \pm 1$  ppm (Fan et al., 2024; Zhang et al., 2024). This paper analyzes cloud top echo signals based on ACDL observation data from July 2022, compares spaceborne IPDA lidar cloud top XCO<sub>2</sub> observations with OCO-2 and CarbonTracker product data, and quantifies ocean carbon absorption intensity. The data comparison and application provide a preliminary analysis of the ability of the IPDA lidar to remotely sense cloud top CO<sub>2</sub> column concentrations.

#### 2. Instrumentation and Methods

#### 2.1. ACDL System and IPDA Lidar

The main mission of ACDL is to use active laser technology for all-weather detection of global atmospheric aerosols and cloud vertical profiles, as well as global atmospheric  $CO_2$  concentration. This provides scientific data for air quality monitoring, studying Earth's carbon cycle, and identifying carbon sources and sinks. The spaceborne ACDL system's laser emits four wavelengths: 532, 1,064, 1,572.024 nm (online), and 1,572.085 nm (offline). The atmospheric  $CO_2$  detection method is the IPDA technique, corresponding to the 1572 nm channel.

#### 2.2. XCO<sub>2</sub> Calculation Methods

The IPDA lidar emits two wavelengths, referred to as the online wavelength and the offline wavelength. As the laser travels along the optical path, the online wavelength is strongly absorbed by  $CO_2$  molecules along the path, while the offline wavelength is only weakly absorbed. By receiving the pulse echo signals of both wavelengths, the  $CO_2$  column concentration along that transmission path is ultimately obtained Figure 1 illustrates the working principle of the spaceborne IPDA lidar system, where  $XCO_2$  can be obtained using Equation 1,

$$XCO_2 = \frac{\tau_{CO_2}}{2 \times 10^{-6} \cdot IWF}$$
(1)

where  $\tau CO_2$  is the differential absorption optical depth (DAOD) and *IWF* denotes the integral weight function (Zhu et al., 2019).





Figure 1. Schematic diagram of spaceborne IPDA lidar principle.

Using the passive satellite OCO-2 and the CarbonTracker (CT) product, it is possible to calculate atmospheric  $XCO_2$  profiles, the results of which can be used for comparison with IPDA lidar cloud top observations. The calculation of the  $XCO_2$  data at the corresponding altitude is represented by Equation 2 (Wunch, D et al., 2010; Mustafa et al., 2020).

$$XCO_{2}^{O} = XCO_{2}^{a} + \sum_{j} P_{j}^{T} K_{j} * (CO_{2}^{i} - CO_{2a})$$
<sup>(2)</sup>

where  $XCO_2^a$  is the a priori value of  $XCO_2$ , P is the pressure weight function, K is the column averaging kernel,  $CO_2^i$  is the CO<sub>2</sub> of the corresponding height stratum of CT,  $CO_{2a}$  is the a priori contour value, *j* is the corresponding vertical height stratum, and *T* stands for matrix transpose.

#### 2.3. Methods for Calculating Ocean Carbon Fluxes

Observations using cloud top  $XCO_2$  can be used to calculate atmospheric ' $XCO_2$ ' below the clouds, and the difference in concentrations can be used to assess changes in carbon fluxes at the subsurface, where in the oceans the carbon uptake can be calculated by Equation 3 (Shi et al., 2021).

$$F_c = \rho w (XCO_{2C} - XCO_{2ABL}) \tag{3}$$

Where  $F_c$  is the ocean carbon uptake,  $\rho$  is the density of the atmospheric boundary layer, w is the vertical wind speed at the cloud top, and  $XCO_{2C}$  and  $XCO_{2ABL}$  are the CO<sub>2</sub> column concentrations at the cloud top and boundary layer, respectively.

# 3. ACDL Cloud Echo Data Analysis and Processing

#### 3.1. Comparison of Altitude Measurement Data

Obtaining accurate cloud top  $XCO_2$  data relies on the precise calculation of the IWF and cloud top pressure, with accurate measurement of cloud top altitude being a key factor (Jacobs et al., 2024). The IPDA lidar system employs a 1,572 nm wavelength in the near-infrared spectrum, achieving sub-meter accuracy in distance measurements (Arnold et al., 2019). This wavelength significantly reduces Rayleigh scattering caused by atmospheric molecules and effectively penetrates atmospheric water vapor, thereby minimizing the impact of atmospheric absorption on measurement precision. Even in the presence of stratus clouds, the system can still attain the necessary accuracy in distance measurements (Gong et al., 2014; Mao et al., 2018; Sharma et al., 2016). The reflectivity of stratus clouds in the 1,572 nm band can be up to 0.05 or even higher, and their surface-hard-target nature can be used as a hard scattering surface for IPDA lidar to obtain accurate cloud top heights (Wang et al., 2020).





Figure 2. Comparison of ACDL 1572 nm channel altitude measurements with SRTM elevation data.

The Shuttle Radar Topography Mission (SRTM), conducted jointly by NASA and the National Geospatial-Intelligence Agency (NGA), is a global elevation measurement project with a global elevation accuracy of 16 m (90% confidence) (Yang et al., 2011). Figure 2 compares ACDL elevation observation data with SRTM data from a region of the African Sahara Desert with minimal human activity and footprints (discontinuities are due to the exclusion of cloud data). In this area, the root mean square error (RMSE) between ACDL and SRTM data is 6.79 m, the coefficient of determination (R<sup>2</sup>) is 0.998, the mean deviation (MD) is approximately 1.24 m, and the standard error (SD) is 6.67 m. These results demonstrate the high reliability of the ACDL system's elevation data.

#### 3.2. Cloud Echoes Data Processing

In the absence of observational equipment with vertical resolution and airborne experiments for comparing cloud top observations, ice crystal particle scattering at higher altitudes in cumulus and cirrus clouds can result in extended optical paths. Significant fluctuations in cumulus cloud top heights and steep gradients of their boundaries can increase observational noise for these cloud types. Additionally, the laser's ability to penetrate most cirrus clouds diminishes detection effectiveness. In contrast, selecting stratus clouds can mitigate the instability of deviations caused by large fluctuations in cloud top height and extended optical paths due to ice crystal particles (Guerlet et al., 2013; Mao et al., 2018, 2024). We use two indicators, echo signal strength and signal-to-noise ratio threshold, to exclude invalid values from different types of echo data.



Figure 3. Peak values and signal-to-noise ratio distributions of echo signals for different types of reflective surfaces. (a) Stratus clouds (b) sea surface (c) ground surface.





Figure 4. Cloud Top, Ground Surface, and Sea Surface Elevation and IWF, DAOD, XCO<sub>2</sub> Distribution (where the Ground examples are a, b, c, and the Sea examples are d, e, f, with shaded areas representing the standard deviation).

According to the statistical comparison results in Figure 3, 93.4%, 97.4%, and 98.8% of stratus clouds top, sea surface, and surface echo signals, respectively, met the screening criteria. This indicates that the three types of echo data exhibit good spatial and temporal continuity. Among them, the land echo is the strongest, followed by the stratus clouds top, while the sea surface is the weakest. As depicted in Figure 3a, the peak distribution of stratus cloud top echo signal intensity ranges from -24 to -27 mV, with the signal-to-noise ratio peaking between 14 and 16. In comparison, sea surface signal intensity peaks between -18 and -20 mV, with a signal-to-noise ratio of 10-11 (Figure 3b). The echo strength from stratus cloud-tops surpasses that of the sea surface, ranking just below ground surface echoes (Figure 3c). This indicates the high quality of stratus cloud top echo signals and the high value of enhancing the utilization of IPDA lidar cloud top data.

## 4. Results

#### 4.1. Calculation of Cloud-Top XCO<sub>2</sub>

The relative variations in both macro- and micro-meteorological phenomena (e.g., wind, convection, and turbulence) and the spatial distribution of surface carbon sources and sinks contribute to fluctuations in  $CO_2$  concentrations influenced by atmospheric dynamics (Abshire et al., 2014; Fu et al., 2018; Lu et al., 2022; Mao et al., 2024). As shown in Figures 4a and 4d, the cloud top heights are approximately 3,695 m and 1,586 m,



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Figure 5. Satellite footprint distribution and vertical observation comparison results.

respectively. The comparison reveals that the cloud top DAOD is smaller, with differences of approximately 0.17 and 0.10. This is due to the higher altitude of the cloud tops compared to the surface and sea level, which reduces the influence of anthropogenic factors and natural emissions. Most  $CO_2$  is concentrated near the surface, leading to higher DAOD from the surface and sea level to the top of the atmosphere compared to that from the cloud top to the top of the atmosphere.

Two of the satellites are about 140 and 165 km apart at their sub-stellar points, as shown in Figure 5. The data from AEMS, OCO-2, and CT exhibit minor differences in the vertical direction, and the trends in vertical variation are consistent, with similar vertical distribution structures. This indicates that the  $CO_2$  concentration distribution in the vertical direction of the atmosphere in this region increases with height. This phenomenon is mainly due to the consumption of  $CO_2$  by daytime terrestrial plant photosynthesis and the absorption of  $CO_2$  by the ocean in the lower atmosphere. Detailed vertical observation comparison results are shown in Table 1. When the signal-to-noise ratio of the cloud top echo is sufficiently high, the accuracy and random error distribution of cloud top XCO2 observations become comparable to those of land echo, exhibiting higher accuracy and reduced random error.

#### 4.2. Assessment of Ocean Carbon Sequestration Capacity

Since the Industrial Revolution, the global oceans have absorbed about 30% of human-emitted  $CO_2$  (Khatiwala et al., 2013). The global oceans' absorption of  $CO_2$  has played a crucial role in mitigating the increase in atmospheric  $CO_2$  levels caused by human activities (Landschützer et al., 2014; Wang, Mustafa, et al., 2021). Furthermore, assessing the amount of  $CO_2$  absorbed by the oceans has always been a key focus and challenge in carbon monitoring. Dang et al. proposed an idealized boundary layer model in 2011 to estimate the regional  $CO_2$  net flux of forests and grasslands. This model has promising applications in regions with minimal human emission interference, such as oceans and forests (Dang et al., 2011). When oceanic  $XCO_{2ABL}$  is calculated to be 392 ppm,

#### Table 1

Comparison of Vertical Observations in Different Data Sets (MRE Represents the Average Random Observation Error, MSNR Stands for Average Echo Signal-To-Noise Ratio)

DataSet	Ground-surf	GS-cloud	Sea-surf	SS-cloud
AEMS	416.8 ± 0.9 ppm	419 ± 1.12 ppm	413.22 ± 1 ppm	413.99 ± 1.5 ppm
	MRE:0.386 ppm (0.096%)	MRE:0.455 ppm (0.11%)	MRE:1.298 ppm (0.32%)	MRE:1.315 ppm (0.33%)
	MSNR:52.8	MSNR:45.9	MSNR:15.5	MSNR:15.8
OCO-2	$417.6 \pm 0.59 \text{ ppm}$	$418 \pm 0.98 \text{ ppm}$	414.9 ± 0.4 ppm	$414.7 \pm 0.6 \text{ ppm}$
СТ	417.78 ppm	419.45 ppm	414.5 ppm	414.6 ppm



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Figure 6. Distribution of ocean carbon absorption.

the oceanic carbon absorption amount for the region is  $-0.319 \text{ mmol/m}^2/\text{h}$  as calculated by Equation 3, with a total uncertainty of 7.2%, aligning well with existing ocean carbon flux data.

As shown in Figure 6, the light blue lines represent the footprints of the AEMS satellite. The carbon absorption amounts in this sea area are -0.26, -0.52, -0.24, and  $-0.32 \text{ mmol/m}^2/\text{h}$  (Figure 6a to Figure 6d), respectively. The small differences between the oceanic carbon absorption amounts calculated by the IPDA lidar and the related data indicate the reliability of calculating oceanic carbon absorption through cloud echo differences. The terrestrial environment exhibits a more complex distribution of anthropogenic carbon emission sources, diverse surface types, and ecosystems. Relevant models are currently under development and will be detailed in future publications.

# 5. Discussion and Conclusions

Due to the high signal-to-noise ratio of the IPDA lidar for certain types of cloud echoes, processing cloud data can significantly enhance the utilization of satellite observation data. In addition, we studied the vertical distribution characteristics of  $CO_2$  based on cloud echoes that can help to identify the carbon sources and sinks in the lower atmosphere and update our dynamic understanding of the carbon cycle.

The IPDA lidar is a highly precise greenhouse gas observation device, but the calculation of the elevation of hard targets is a critical factor that is affecting its accuracy. By comparing with SRTM data, the ACDL elevation data achieve an  $R^2$  of 0.998 with a root mean square error of only 6.67 m. The AEMS cloud top XCO<sub>2</sub> results show small differences from those of passive satellites and models, with similar trends. The oceanic carbon absorption capability was assessed using XCO<sub>2</sub> concentration differences based on cloud echo observations from the spaceborne IPDA lidar and vertical height layers at the sea surface. The results show that the ocean carbon absorption in the region is  $-0.319 \text{ mmol/m}^2/\text{h}$ , which is consistent with carbon flux data from GCB, CMS, CT, and CMEMS-LSCE. Future work will involve a detailed analysis of other types of cloud echoes and will use nighttime lidar observation data to estimate diurnal and nocturnal ocean carbon absorption, improving the application of data in scientific research.

# **Conflict of Interest**

The authors declare no conflicts of interest relevant to this study.

#### **Data Availability Statement**

SRTM global elevation data can be accessed at https://srtm.csi.cgiar.org/srtmdata/. The  $CO_2$  profile data provided by OCO-2 can be obtained from (OCO-2/OCO-3 Science Team et al., 2020). Vertical wind speed data from ERA5 (Hersbach et al., 2023). Ocean carbon flux data can be obtained from Global Carbon Budget (Hauck et al., 2023), Carbon Monitoring System (Liu & Bowman, 2024), CarbonTracker (Jacobson et al., 2024), and Copernicus Marine Service (Global Ocean Surface Carbon, 2023; Chau, T.-T.-T et al., 2022). The AEMS data used in this study was not publicly available at the time of submission. The data can be requested at https://data. cresda.cn/#/home but is not accessible outside of China. Researchers in China with relevant licenses have free access to satellite data.

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